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Recording Radiographic Images on Nitrocellulose Film in Neutron Radiography of Nuclear Reactor Fuel

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August 1987

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RECORDING RADIOGRAPHIC IMAGES ON NITROCELLULOSE FILM IN NEUTRON RADIOGRAPHY OF NUCLEAR REACTOR FUEL

J.C. Domanus

Abstract. Nitrocellulose film, insensitive to X- and gamma-rays (as well as visible light), is especially suitable for neutron radiography of spent nuclear reactor fuel, which itself is a source of high-intensity gamma-radiation. As nitrocellulose film is also insensitive to neutrons a converter is necessary to convert neutrons to alpha particles, which in turn are able to produce a radiographic image on the film. This image, in the form of minute pits in the film, must thereafter be made visible by etching the nitrocellulose film to thereby enlarge the pits so much as to produce a visible image on the film. After a short description of neutron radiography facilities at various reactor types three methods of producing neutron radiographs are explained (direct, transfer and track-etch). Nitrocellulose film and neutron-to-alpha converters used with it are described. Radiographic image quality of this film is compared with that of silver halide film. A similar comparison is made of the accuracy of dimensional measurements from neutron radiographs.

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1. INTRODUCTION

There are two main reasons why nitrocellulose film is used in neutron radiography.

- It can be used as a direct-imaging technique for radioactive objects.
- It produces sharper images than the silver halide film.

Whereas the first advantage of using nitrocellulose film is self-evident (as the film is insensitive to the radiation emitted by the radioactive object) the second advantage is still to be proved.

As in other fields of industrial radiography image quality indicators (IQI) are also used in neutron radiography to assess the quality of the radiographic image. Up till now such IQIs for neutron radiography were published in one ASTM (1975, 1981, 1986) and one AFNOR (1982) standards. The 1975 and 1981 issues of the ASTM were designed only for silver halide film and the 1986 issue further restricted the use of the standard to the direct method. The AFNOR standard (1982) is designed for non radioactive objects.

The ASTM (1986) standard describes two types of IQIs: the Beam Purity Indicator (BPI) (which is restricted to only single-coated silver halide film and the direct method) and the Sensitivity Indicator (SI).

As none of those standards was intended for use with nuclear reactor fuel the Euratom Neutron Radiography Working Group (NRWG) has designed and produced two special indicators for that purpose: The Beam Purity Indicator-Fuel (BPI-F) and the Calibration Fuel Pin (CFP-E1). Those two indicators, together with the ASTM Beam Purity Indicator (BPI) and Sensitivity Indicator (SI) were tested by all the participants of the NRWG under a special test program.

One of the aims of this test program is to investigate whether the BPI, BPI-F and SI can be used with nitrocellulose film.

The second aim is to test the accuracy of measuring dimensions from neutron radiographs made on nitrocellulose film.

2. PRINCIPLES OF NEUTRON RADIOGRAPHY

All radiographic methods, whether they make use of X-rays, gamma-rays or neutrons, are based on the same general principle: that radiatiaon is attenuated on passing through matter.

Thus, the detection of defects in radiography is based on the observation of differences in radiation intensity after passing through the object under examination. This occurs according to the basic law of radiation attenuation:

$$J = J_o \, e^{-\mu x}$$

The radiation attenuation coefficient μ shows a continous curve for X-rays (over a wide range of wavelengths). This is not the case, however, for neutrons and it happens that adjacent atomic number elements such as boron and carbon show for example marked differences in neutron attenuation. Because of this it is possible to detect hydrogen in zirconium. Conversely, dense materials such as lead, tungsten, or uranium are relatively transparent to neutrons. Another important advantage of neutron radiography is its ability to examine directly radioactive objects such as spent fuel elements.

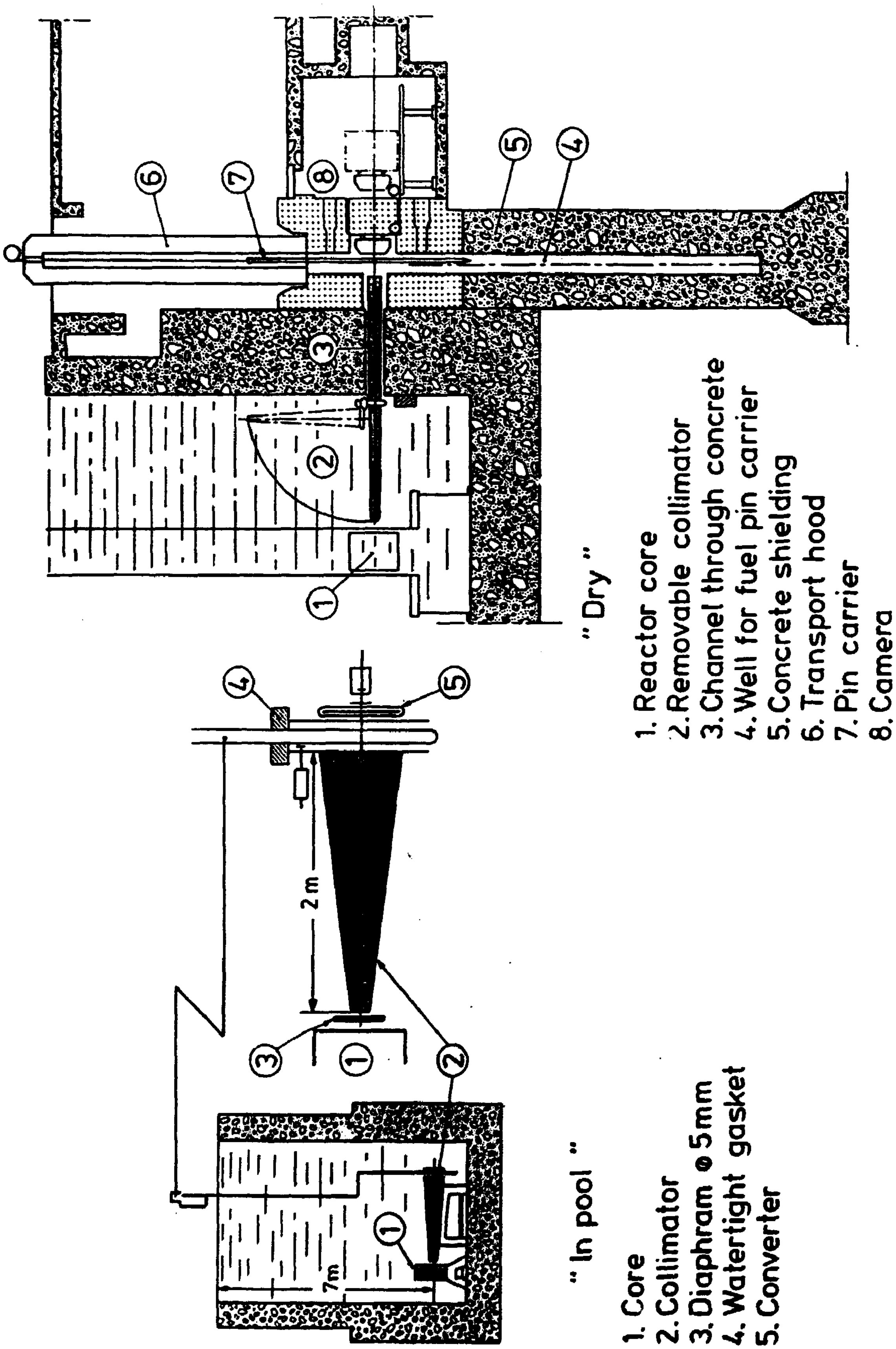

Fig. 1. "In pool" and "dry" reactor facility.

3.RADIATION SOURCES

There are three sources of neutrons available for neutron radiography: particle accelerators, radioisotopes and nuclear reactors. Only the latter will be reviewed below. At present nuclear reactors provide the most intense neutron beams and therefore can produce neutron radiographs of the highest quality.

Two types of neutron radiographic facilities are used with nuclear reactors (see fig. 1). In the "in pool" facility the whole neutron radiographic installation is immersed in the pool of the reactor. Here, irradiated reactor fuel rods, removed from the reactor core, are transferred to the neutron radiographic facility, where they are examined without removing them from the reactor pool.

In the "dry" type facility, a neutron beam taken out of the core of the reactor is used outside the reactor for neutron radiography.

4. NEUTRON RADIOGRAPHIC TECHNIQUES

As in X- or gamma-radiography, X-ray film is the medium for producing neutron radiographs. For radiographing radioactive materials the nitrocellulose film is also used.

Unfortunately, neutrons have very little direct effect on photographic film. Thus an intensifying screen of some kind is needed to increase the speed of the film. The nitrocellulose film must also be used with a converter screen, as neutrons do not directly affect this type of film.

Of the many existing methods of recording neutron images, only those which are widely used in practice will be described here (see fig. 2). They are the following: the direct and transfer technique using metal converter foils with X-ray film and the track-etch technique using nitrocellulose film.

4.1. Direct Exposure Technique. In the direct exposure technique a metal converter foil is placed in contact with X-ray film during the actual exposure (fig. 2). Usually, a single gadolinium back screen is used. This screen emits gamma-radiation on absorbing neutrons. The gammas in the spectrum from gadolinium are suitable for producing electrons by internal conversion. Those low-energy electrons essentially expose only the emulsion facing the gadolinium. Single coated slow X-ray films are therefore used with the direct technique.

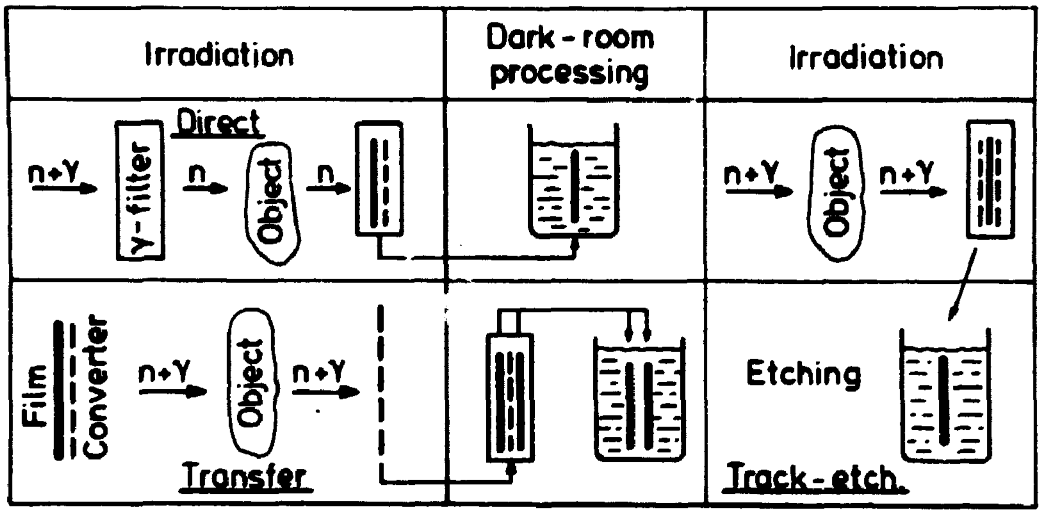

Fig. 2. Neutron radiographic techniques.

4.2. Transfer Technique. In the transfer technique (fig. 2) only the converter screen is exposed directly to neutrons. The metal screen placed in the neutron beam becomes radioactive in proportion to the intensities in the spatial neutron image. The screen is subsequently transferred from the neutron beam to a darkroom where it is placed in close contact with the X-ray film using a vacuum cassette. The radioactive emission from the screen then produces an image on the film.

The transfer method offers the advantage that the film, which is not present in the neutron beam, is neither exposed to gamma-radiation from a radioactive object nor from gamma-rays in the neutron beam itself.

The more energetic β-particles emitted by the transfer screens have sufficient energy to penetrate normal photosensitive film completely. Thus both emulsions of a double coated X-ray film will contribute appreciably to the film density and to the unsharpness.

4.3. Track-etch Technique. For neutron radiography of radioactive objects (such as irradiated nuclear fuel) nitrocellulose film is used as a neutron detector. This is a dielectric material which can detect charged particles by the radiation damage caused in it. Those charged particles are produced by an α-emitting converter screen. The radiation damage is made visible by etching in hot sodium hydroxide solution (e.g., in 10% NaOH for 45 min at 50°C). The nitrocellulose film, sandwiched between two α-emitting converter screens, is placed directly in the neutron beam (fig. 2) as it is insensitive to gamma-rays. As the nitrocellulose film is also insensitive to visible light the consecutive etching need not be done in a dark room.

Nitrocellulose film is available (from Kodak-Pathé, France) for neutron radiography in two film/converter variations. The first consists of a 100 μm thick sheet of cellulose nitrate coated on both sides with lithium borate dispersed in a water-soluble binder, which acts as a converter screen by means of the (n,α) reaction (CN 85 Type B). After irradiation the lithium borate coating is removed by washing and then the film itself is etched.

The second variation consists of the same CN 85 nitrocellulose film (without coating) which is sandwiched between two converter screens (BN 1) made from natural boron, a (n,α) converter. This converter is coated on a 100 μm thick, very stable, polyester base and can be reused indefinitely. The efficiency of the BN1 is higher than that of the CN 85 Type B and therefore requires exposure times only slightly longer than those for the transfer technique (with Dy converter and slow X-ray film). To establish perfect contact between the converter and nitrocellulose film the use of a vacuum cassette is essential.

5. PROCESSING OF NITROCELLULOSE FILM

Unlike that of the silver halide film the processing of nitrocellulose film can be adapted to the object being radiographed. The processing consists of the etching of the exposed nitrocellulose film in a solution (of various concentrations) of the etching agent at different time/temperature combinations. The most commonly used etching agent is the 10% (2.5 N) solution of analytical grade sodium hydroxide in distilled water. As recommended by the film manufacturer the processing conditions may vary from 10 to 30 min at 60°C to 2 to 8 h at 25°C (during the NRWG Test Program the films were etched at 20°C for 21 h and at 50°C for 45 min). In general when a low-concentration, low-temperature or short etching time is adopted then those parts on the neutron radiographs that have the low attenuation of neutrons will appear. For longer etching times, however, those parts with the higher neutron attenuation will appear more clearly. This permits the use of the so called gradual processing, where the etching will first be stopped after a shorter time and the radiograph will be viewed to see the parts of lower attenuation, and then the etching will continue until it reaches the time necessary to see the parts that have the highest attenuation of neutrons.

6. VIEWING OF NEUTRON RADIOGRAPHS

The viewing of neutron radiographs produced by the direct or transfer method on X-ray film creates no special problems.

As the optical density of the etched nitrocellulose film is rather low compared with the film densities of silver halide films used in X- and gamma radiography, two additional methods of viewing neutron radiographs on nitrocellulose film (besides direct viewing) are used to increase the low contrast of the radiographic image.

The contrast can be significantly improved by printing the nitrocellulose film on high contrast film, using a point source enlarger. However, the nitrocellulose film can be directly examined by placing it between two polarizing filters (for direct viewing).

7. RADIOGRAPHIC IMAGE QUALITY

One of the most important factors that is always checked in industrial radiography is the radiographic image quality. Therefore it will be best to compare the nitrocellulose with the silver halide film by investigating image quality of both. This could be done by the use of image quality indicators (IQI). Unfortunately, such IQIs do not exist for neutron radiography performed on nitrocellulose film.

Under the NRWG Test Program the ASTM E545 SI was used as an IQI.

The examination of about 150 neutron radiographs of the SI has shown that in more than 96.5% all the Al shims of the SI were seen on all neutron radiographs. Therefore the examination of the visibility of Al shims on neutron radiographs is not selective enough to compare the image quality of nitrocellulose and silver halide film.

The same examination performed about the visibility of holes on the same 150 neutron radiographs of the SI has shown that more holes can almost always be seen on silver halide than on nitrocellulose film. Only on films exposed on one of the NR facilities could more holes be seen on nitrocellulose film.

Closing the above remarks about the assessment of the image quality of nitrocellulose film by the use of the ASTM SI one must stress once more that the ASTM standard was designed only for silver halide film.

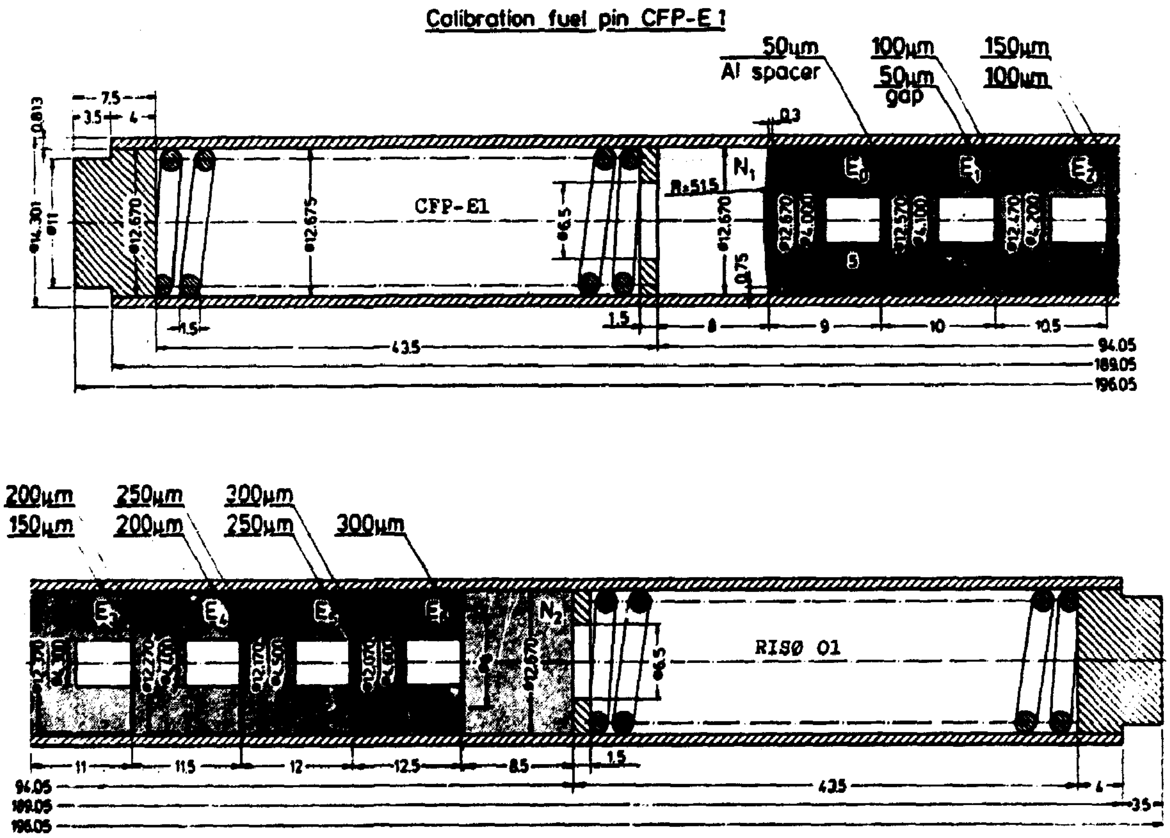

Fig. 3. Calibration fuel pin CFP-E1.

Following the statement of the ASTM E545 "that the only truly valid sensitivity indicator is a material or component, equivalent to the part being neutron radiographed, with a known standard discontinuity (reference standard comparison part)" a calibration fuel pin (CFP) was designed and produced at Risø (see fig. 3). The first version of this CFP was used as early as in 1976 to check the accuracy of dimensional measurements from neutron radiographs of nuclear fuel pins (Domanus, 1976).

This CFP was thereafter used also for comparing the image quality of nuclear fuel neutron radiographs taken on silver-halide film and the CA80-15B and CN85B nitrocellulose films (Domanus, 1979).

Finally a similar comparison (using the CN85B film) was also reported (Domanus, 1981).

There are several methods to use the CFP for the purpose of assessing the image quality of neutron radiographs. CFP was especially designed to assess neutron radiographs of nuclear fuel. This is just the field in which nitrocellulose film is most useful and mostly utilized.

Viewing a neutron radiograph of a CFP one can assess its quality visually. This was previously done and reported using a five grade arbitrary scale of image quality. The highest grade 5 was used, when a neutron radiograph of the CFP was equally sharp as an X-ray radiograph of the samea CFP. The poorest neutron radiographs were given the grade of 1.

The former examinations were limited to only one NR facility (Risø DR1) with a rather low L/D = 110.

Now a larger experimental material is available in the form of about 150 neutron radiographs originating from 11 NR facilities with different L/D ratios. The results of this examination were reported by Domanus (1987).

8. ACCURACY OF DIMENSIONAL MEASUREMENTS

The second method of using the neutron radiographs of the CFP-E1 for assessing image quality consists of making actual measurements from neutron radiographs of different dimensions of the CFP-E1 and comparing them with the true dimensions given in CFP-E1 measuring certificate. From this comparison conclusions can be drawn about the image quality of silver halide as well as nitrocellulose film.

The dimensions can be measured using either a projection microscope or a travelling microdensitometer. In the first instance a subjective measuring method is in use whereas in the second objective results are obtained.

During the NRWG Test Program about 25,000 dimensional measurements were made. At eleven neutron radiographic facilities the CFP-E1 was neutron radiographed and the radiographs were processed and assessed 402 times (273 times with the profile projector and 129 with the travelling microdensitometer) using 30 different recording and viewing techniques. The results of those measurements are now evaluated in three groups by calculating standard deviations between the measured and true dimensions:

1) For all the eleven neutron radiography facilities together.
2) For each facility individually.
3) For different groups of dimensions (e.g. pellet diameter, pellet length, pellet-to-pellet and pellet-to-clad gaps).

As the results of this evaluation are as yet unavailable, only preliminary conclusions can be drawn about the accuracy of dimensional measurements from neutron radiographs.

9. CONCLUSIONS

When comparing radiographic image quality and accuracy of dimensional measurements of the nitrocellulose film with those of the silver halide film one must remember that the main field of use of the former is neutron radiography of radioactive objects (such as irradiated nuclear reactor fuel). Therefore the comparison ought to be done between the track-etch method using nitrocellulose film, and the transfer method using silver halide film.

9.1. Image quality. When analyzing the results of the image quality assessment by the subjective (visual) method one can come to the following conclusions:

- in many instances the quality of the neutron radiographs taken on nitrocellulose film was equally good as that on a single-coated silver halide film (exposed by the transfer method using Dy converter), but it was never better;
- in several instances the quality of the neutron radiographs taken on silver halide film by the transfer method was better than that of the nitrocellulose film;
- the use of polarizing filters for the visual assessment of image quality of the nitrocellulose film did not improve the image quality; sometimes the quality was equally good as without the polarizing filters, but never better;
- in several instances the quality was better for CN85 type B films (coated with converter on both sides) than for CN85 film sandwiched between two BN1 converters; only in one instance was the reverse true.
- in several instances etching at 20°C for 21 h gave better quality than etching at 50°C for 45 min.

As a general conclusion from the visual assessment of radiographic quality one may state that nitrocellullose film is no better than single-coated silver halide film used with the Dy converter and the transfer method.

9.2. Dimensional measurements. From the partial analysis of results obtained from same dimensional measurements (gaps) of the CFP-E1 one can come to the following conclusions:

- almost always the percent deviation between the measured and true dimensions is much greater for the small gap (50 μm) than for the large one (300 μm),
- for the large gap (300 μm) the largest deviation was 35% and the smallest 0.3%,
- when calculating the above deviations in absolute figures one will have for the smallest gap the largest deviation 127.5 μm and the smallest deviation of 1 μm, whereas for the largest gap the corresponding results will be 105 and 0.9 μm,
- most of the measurements have shown larger values than the true dimensions,
- as could be expected the best results among the silver halide films were almost always obtained with the single-coated Kodak SR film,
- in many instances nitrocellulose films etched at 20°C for 21 h gave better results than those etched at 50°C for 45 min; however, this cannot be accepted as a rule and therefore one cannot recommend the use of the one or the other mode of etching,
- as for the use of polarizing filters, in many instances the measuring accuracies obtained with them were better than without them, but the difference was not so convincing as to justify the recommendation of using polarizing filters for dimensional measurements from nitrocellulose film,
- in general the use of CN85 type B films (coated with converter on both sides) gave better measuring accuracies than those obtained with the CN85 film sandwiched between two BN1 converters; one must remember, however, that this latter combination permits shorter exposure times,
- in some instances measuring accuracies obtained with nitrocellulose film were better than those with the best (single coated) silver halide film; the accuracies were not, however, so spectacular as one could expect,

- one could not find a direct relationship between the accuracy of dimensional measurements and the L/D ratio of different NR facilities.

As a general conclusion from the above measuring results one can say that one cannot recommend the use of nitrocellulose film based only on their ability to give occasionally higher accuracies in dimensional measurements.

10. ADDITIONAL INFORMATION

Additional information about nitrocellulose film can be found in a special publication dedicated to that subject. It is the EUR report "Neutron Radiography on Nitrocellulose Film", edited by J. Markgraf. It will be published in 1988 as a book by the D. Reidel Publishing Co. in their series on neutron radiography.

Many examples of neutron radiographys of nuclear reactor fuel taken on nitrocellulose film can be found in another EUR report (published in 1984) by D. Reidel in the series on neutron radiography): "Reference Neutron Radiographs of Nuclear Reactor Fuel", edited by J.C. Domanus.

REFERENCES

AFNOR (1982). Norme experimentale A09-220. Neutronographie industrielle. Determination des characteristiques des installations pour controle de pieces non radioactives. Industrial neutron radiography. Determination of the characteristics of testing facilities for the control of nonradioactive parts. Septembre 1982.

ASTM E545-75. Standard method for determining image quality in thermal neutron radiographic testing.

ASTM E545-81. Standard method for determining image quality in thermal neutron radiographic testing.

ASTM E545-86. Standard method for determining image quality in direct ther mal neutron radiographic testing.

Domanus, J.C., (1976) Accuracy of dimension measurements from neutron radiographs of nuclear fuel pins. Risø-M-1860. 23.03.1976 (also as paper 3L8 presented at the 8th World Conference on Nondestructive Testing, Cannes, 6-11.9.1976).

Domanus, J.C., (1979) Comparison of image quality of nuclear fuel neutron radiographs taken on silver halide and nitrocellulose film. Risø-M-2170. April 1979 (also as paper 2BDD-1 of the 9th World Conference on Non-destructive Testing, Melbourne, 18-23.11.1979).

Domanus, J.C., (1981) How good is nitrocellulose film for neutron radiography? <u>Neutron Radiography, Proc. First World Conference</u>. San Diego, 7-10.12.1981. D. Reidel Publishing Co., 729-736.

Domanus, J.C., (editor) (1984). <u>Reference neutron radiographs of nuclear reactor fuel</u>. D. Reidel Publishing Co., Dordrecht/ Boston/Lancaster.

Markgraf, J. (editor) (to be published). <u>Neutron radiography on nitrocellu lose film</u>. D. Reidel Publishing Co., Dordrecht/Boston/London.

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